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Time-Dependent Simulations of Laminar Flames in Hydrogen-Air Mixtures

K. KAILASANATH AND E. S. ORAN

Laboratory for Computational Physics and Fluid Dynamics

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TIME-DEPENDENT SIMULATIONS OF LAMINAR FLAMES IN HYDROGEN-AIR MIXTURES

1. Introduction

In this paper, we present results of detailed numerical simulations of laminar flames in hydrogen-air mixtures. We use these simulations to discuss two major problems: (1) the relative importance of various diffusive transport processes on flame propagation in fuel-lean, stoichiometric, and fuel-rich mixtures, and (2) the time-dependent behavior of flames near the rich flammability limit.

Various numerical simulation techniques have been applied to study a wide variety of problems involving the ignition, propagation and quenching of hydrogen flames. However, there have not been many attempts to use the detailed modeling approach to study quantitatively the relative importance of various physical mechanisms involved in flame propagation. In one such effort [1], numerical simulations were used to evaluate the relative importance of thermal conduction and diffusion of species in specific methane and methanol flames. The calculations seemed to indicate that flame propagation is not governed primarily by diffusion of radicals but diffusion of fuel which subsequently affects the chemical reactions. More recently, we [2] showed that neither diffusion of species nor thermal conduction alone could give the correct burning velocity for a stoichiometric hydrogen-oxygen-nitrogen mixture. In the first part of this paper, we present a systematic study of the relative importance of various diffusive transport processes to flame propagation in fuel-lean, stoichiometric and fuel-rich hydrogen-air mixtures.

In an earlier study on the effects of curvature and dilution on flame propagation [3], we showed that a flame can be extinguished with less dilution in one geometry than in another, even in the absence of external heat losses and buoyancy effects. Specifically, we showed that a planar flame can propagate steadily in a dilute mixture which does not support a spherically expanding flame. Because this behavior is related to the effects of flame stretch, it raises the fundamental question of whether there is an extinction limit in the absence of stretch effects. This question can be addressed by carefully studying the behavior of planar flames in the absence of external heat losses and gravitational effects. Both lean and dilute hydrogen-oxygen-nitrogen mixtures are subject to flame instabilities.

Therefore, we restrict ourselves to the extinguishment behavior of fuel-rich hydrogen-air flames in the second part of this paper.

2. The Numerical Model

The results presented in this paper are derived from numerical simulations using FLAME1D, a one-dimensional, Lagrangian model [4,5]. The model solves the time-dependent conservation equations for mass, momentum and energy [6,7]. The model has been used for a variety of flame studies, including calculations of burning velocities [5], minimum ignition energies [8] and quench volumes [8]. The model has a modular form and permits a wide variety of geometric, initial, boundary, and time varying energy input conditions.

The convective transport is solved by ADINC, a Lagrangian algorithm which solves implicitly for the pressure [9]. Because the method is Lagrangian and there is no numerical diffusion due to convective transport, the method gives an accurate representation of material interfaces and allows steep gradients in species and temperature to develop and be maintained. An adaptive Lagrangian regridding algorithm refines the grid ahead of the flame-front so that the flame zone remains well resolved throughout the calculation. Thermal conduction, ordinary diffusion, and thermal diffusion processes are included for each chemical species. The chemical interactions are described by a set of nonlinear, coupled ordinary differential equations which are solved using a fully vectorized version of the selected asymptotic integration method CHEMEO [10,11]. The algorithms representing the various chemical and physical processes are integrated separately and then asymptotically coupled by timestep-splitting techniques [7].

For this study we have used the hydrogen-oxygen reaction scheme [12] which involves the eight reactive species H_2 , O_2 , H , O , OH , H_2O , HO_2 , H_2O_2 , and the diluent, N_2 . The thermochemical properties of the various species involved are taken from the JANAF tables [13]. The chemical reaction mechanism [4,8,12], consisting of about fifty steps has been extensively tested and shown to give good results. Burks and Oran [12] showed that the results computed with this mechanism compared well with experimentally observed induction times, second explosion limits and the temporal behavior of reactive species. Oran et al. [14] have shown that when coupled to a convective transport algorithm, the mechanism gives good results in the simulation of the conditions behind a reflected shock. The laminar burning velocities calculated using the mechanism are in agreement with experimental data [5].

In the calculations presented below, the model was configured with an open boundary at one end to simulate an unconfined system. All the calculations were performed in a planar

(Cartesian) geometry. The initial temperature and pressure of the mixtures considered were 298 K and 1 atm, respectively.

The calculations were initialized with a Gaussian temperature profile. The central temperature and width were chosen so that the added initial energy was above the minimum ignition energy for the mixture. That is, it was ensured that the initial conditions provided enough energy for the flame to evolve to a steady profile.

For either thin or planar flames, the instantaneous normal burning velocity can be calculated from the flame velocity if we know the velocity of the unburnt gases ahead of the flame. As shown in Fig. 1, for planar flames the velocity of the unburnt gases ahead of the flame is constant. Hence the burning velocity can be unambiguously determined as the difference between the flame velocity and the flow velocity ahead of the flame.

$$V_{burn} = V_{flame} - V_{fluid}$$

We have discussed this definition and its application to curved flames elsewhere [3].

3. Relative Effects of Various Diffusive Transport Processes

A series of calculations have been performed to study the effects of the individual diffusive transport processes on flame propagation in hydrogen-air mixtures. The three different diffusive transport processes we consider are 1) thermal conduction, 2) ordinary diffusion, and 3) thermal diffusion. In order to isolate the relative importance of each, we have performed calculations with different combinations of these processes turned on. We repeated the same sequence of calculations for three mixtures:

Fuel-lean: H₂:O₂:N₂ / 0.65:1:3.76

Stoichiometric: H₂:O₂:N₂ / 2:1:3.76

Fuel-rich: H₂:O₂:N₂ / 15:1:3.76.

Table 1 summarizes the calculated burning velocities from this series of calculations. For each of the three mixtures considered we show four different cases:

- (a) All three diffusive transport processes are on;
- (b) Thermal diffusion is off, and thermal conduction and ordinary diffusion are on;
- (c) Thermal conduction is on, and thermal diffusion and ordinary diffusion are off;
- (d) Thermal conduction off, and ordinary and thermal diffusion are on.

Several interesting trends in burning velocities appear from looking at this table. First, we see that the relative importance of thermal diffusion does not change from lean to rich mixtures. However, the relative importance of thermal conduction and ordinary diffusion are different for lean, stoichiometric, and rich mixtures.

3.1 The Effect of Thermal Diffusion on Burning Velocities

By comparing cases (a) and (b) in Table 1, we see that the effect of thermal diffusion is to lower the burning velocities by 6-10% in all three mixtures. The effects of thermal diffusion in hydrogen-air mixtures have been studied earlier [4,15,16].

Hydrogen atoms are the fastest moving, most easily diffused species in the system. Their diffusion behavior dominates the diffusion properties of the hydrogen-air system. Hydrogen atoms are formed in the high-temperature region of the flamefront, and are then diffused into the cold mixture by ordinary diffusion. Earlier studies have shown that thermal diffusion tends to diffuse hydrogen atoms from the lower temperature region to the higher temperature region [4,15]. Thus these two processes have the opposite effect with respect to moving hydrogen atoms. The increase in burning velocity when there is no thermal diffusion occurs because thermal diffusion inhibits the diffusion of hydrogen radicals into the cold, unburnt mixture ahead of the flamefront.

3.2 Stoichiometric Mixture

For the stoichiometric mixture, thermal conduction and ordinary diffusion are almost equally important in determining the burning velocity of the flame. Without thermal conduction, the burning velocity is reduced to 62% of its standard value, i.e., the value of the burning velocity with all of the diffusive transport processes turned on. Without ordinary diffusion, it is reduced to 46% of its standard value. Figure 2 shows the behavior of the flame and burning velocities as a function of time. The initial condition causes a large initial velocity which eventually relaxes to a steady flame velocity.

3.3 Fuel-Lean Mixture

For the lean mixture, Table 1 shows that turning off either thermal conduction or ordinary diffusion lowers the burning velocity. In particular, we note that thermal conduction is much more important than ordinary diffusion in determining whether or not the flame propagates. When the thermal conduction is turned off, the flame does not reach a steady burning velocity at all.

Figure 3 shows the calculated flame velocity and fluid velocity as a function of time for case (d), where the flame appears to die. The flame velocity decreases with time, and becomes practically identical to the fluid velocity. The result is a negligible burning velocity. Using a time-dependent model to study burning velocities allows us to see the flame die out in time. No changes in the initial conditions which we tried could keep this flame from eventually "dying out" in the way shown in the figure.

For this lean flame, radicals can diffuse ahead of the flame front, but without thermal conduction, the temperature ahead of the flame never becomes high enough to sustain the reaction. The fuel deficit also means a deficit of hydrogen radicals, which hinders the reaction process. Thermal conduction alone, however, carries forward enough heat to create new radicals and sustain the process. Figure 4 shows the difference in the hydrogen radical profiles for cases (a), (c) and (d) at typical times in the calculation.

3.4 Fuel-Rich Mixture

For the fuel-rich mixture, ordinary diffusion is the most important process in determining the burning velocity. In contrast to the fuel-lean flame, the fuel-rich flame does not propagate without ordinary diffusion. In the fuel-rich case, not enough heat can be transferred by conduction alone to sustain the reactions. However, because of the excess of hydrogen radicals, ordinary diffusion alone can spread enough radicals ahead of the front to initiate chemical reactions and sustain the flame.

4. Flames in Fuel-rich Mixtures

In the second set of calculations we restricted ourselves to flames in fuel-rich hydrogen-air mixtures. In these calculations all the diffusive transport processes discussed above were included. The amount of hydrogen in the mixture was systematically increased and with each concentration of hydrogen, the calculations were carried out until a steadily propagating flame (if any) was observed. The burning velocities were calculated as discussed earlier. The burning velocity and the temperature of the burned gases (flame temperature) for the various mixtures are summarized in Table 2. As expected, the burning velocity and the flame temperature decrease with increasing fuel concentration. However, a couple of interesting observations can be made from the data presented in the table.

First of all, for a 80.8% hydrogen-air mixture ($H_2:O_2:N_2 / 20:1:3.76$), we obtain a steady burning velocity of about 8 cm/s. This mixture is beyond the experimentally observed rich-flammability limit under normal gravity conditions. This difference between the numerical and experimental observations may be due to the effects of gravity, heat losses to the confining walls in the experiments or multidimensional effects. As mentioned earlier, these effects have not been included in these numerical simulations. For example, we know from previous calculations [3] that curvature (or stretch) can reduce the burning velocity.

The second interesting observation from the numerical simulations is that a 82.2% hydrogen-air mixture ($H_2:O_2:N_2 / 22:1:3.76$) does not support a steadily propagating flame. We tried different ignition sources but in all cases the burning velocity decreased to zero. The behavior of the transient flame was qualitatively similar to that shown in Fig. 3 for a dying flame. The simulations indicate that there is a flammability limit even in the absence

of stretch, buoyancy and external heat losses. Furthermore this limit is higher than that observed in the standard flammability limit tube under normal gravity conditions.

A possible reason for this flammability limit can be seen in the trend of flame temperatures shown in Table 2. The flame temperature for a 20:1:3.76 mixture is just 970 K. With increase in fuel concentration the temperature would decrease further. At these temperatures, it is possible that the H atoms are consumed faster in chain terminating reactions than produced in chain branching reactions. This would result in a depletion of H atoms and a decrease in temperature. This could eventually lead to a situation where the energy released in exothermic reactions is not sufficient to balance the energy diffused by the transport processes and hence a steady flame is not possible. This possibility is currently being investigated.

5. Summary and Conclusions

We have examined two fundamental problems in premixed laminar flames using a detailed one-dimensional model. The model is time-dependent and can calculate the evolution to a steady-state, propagating flame from an initial energy source. Because it is time-dependent, the absence of a steady-state can also be revealed by it.

In the first problem, we examined the relative importance of thermal conduction, thermal diffusion, and ordinary diffusion in determining the burning velocities of laminar hydrogen-air flames. Three cases were examined in detail: a fuel-lean mixture, a stoichiometric mixture, and a fuel-rich mixture. For the fuel-lean and fuel-rich mixtures, care was taken to ensure that the mixture considered was far enough from the limit to have a finite, but relatively low, burning velocity. The results are summarized in Table 1, which shows the burning velocities calculated for the three mixtures.

The effect of thermal diffusion is to decrease the burning velocity by 5-10%, in agreement with previous calculations [4,15,16]. This can be explained by noting that the effect of thermal diffusion is to diffuse hydrogen radicals in towards the hot region of the flame, instead of out towards the unburned region. It has the opposite effect of ordinary diffusion and thus inhibits the flame-propagation process.

For the stoichiometric mixture, both thermal conduction and ordinary diffusion make important contributions to sustaining the flame. When thermal conduction was turned off, the burning velocity was reduced to 62% of its standard value. When ordinary diffusion was turned off, the burning velocity decreased to 46% of its standard value. These results were qualitatively as expected.

The more surprising results were for the fuel-rich and the fuel-lean mixtures. For the fuel-lean mixture, the flame would not propagate when thermal conduction was turned off.

That is, ordinary diffusion alone could not sustain the flame. However, the burning velocity was only 69% of its standard value with just thermal conduction.

In contrast to the fuel-lean behavior, the fuel-rich mixture could not sustain a flame when ordinary diffusion was turned off. That is, thermal conduction alone could not sustain the flame. With just ordinary and thermal diffusion, the burning velocity was 86% of its standard value.

Our general conclusion from this study is that both ordinary diffusion and thermal conduction are necessary to quantitatively describe flame propagation in a hydrogen-air mixture. Their relative importance, however, varies as we go from fuel-lean to fuel-rich hydrogen-air mixtures. It is not clear that these conclusions hold for hydrocarbon combustion, since there are other, heavier radicals that might change the balance between thermal conduction and ordinary diffusion. An implication of this work to flame modelling efforts is that it may be possible to neglect thermal diffusion, but both molecular diffusion and thermal conduction must be included in the model.

In the second part of this paper we considered the behavior of flames in fuel rich hydrogen-air mixtures near the experimentally observed flammability limit. The effects of gravity, stretch and external heat losses were eliminated in the numerical simulations. The results suggest wider flammability limits than those observed experimentally under normal gravity conditions. The simulations indicate that there may be a limit due to chemical kinetic considerations alone. Further details of a possible chemical-kinetic limit are currently under investigation. The simulations also suggest that multi-dimensional calculations under normal gravity conditions and experiments under reduced gravity conditions are needed to better understand flames near the standard flammability limits.

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Table 1. - Burning Velocities of H₂-Air Mixtures

Case	Processes On	Lean	Stoichiometric	Rich
(a)	All	36 cm/s	185 cm/s	44 cm/s
(b)	Thermal Conduction Ordinary Diffusion	40	195	48
(c)	Thermal Conduction	25	85	0
(d)	Ordinary Diffusion Thermal Diffusion	0	115	38

Table 2. - Flames in Fuel-Rich Mixtures

Mixture	Burning Velocity	Flame Temperature
H ₂ : O ₂ : N ₂	(cm/s)	(K)
16:1:3.76	38	1180
17:1:3.76	30	1108
18:1:3.76	23	1046
19:1:3.76	16	1002
20:1:3.76	8	970
22:1:3.76	—	<970

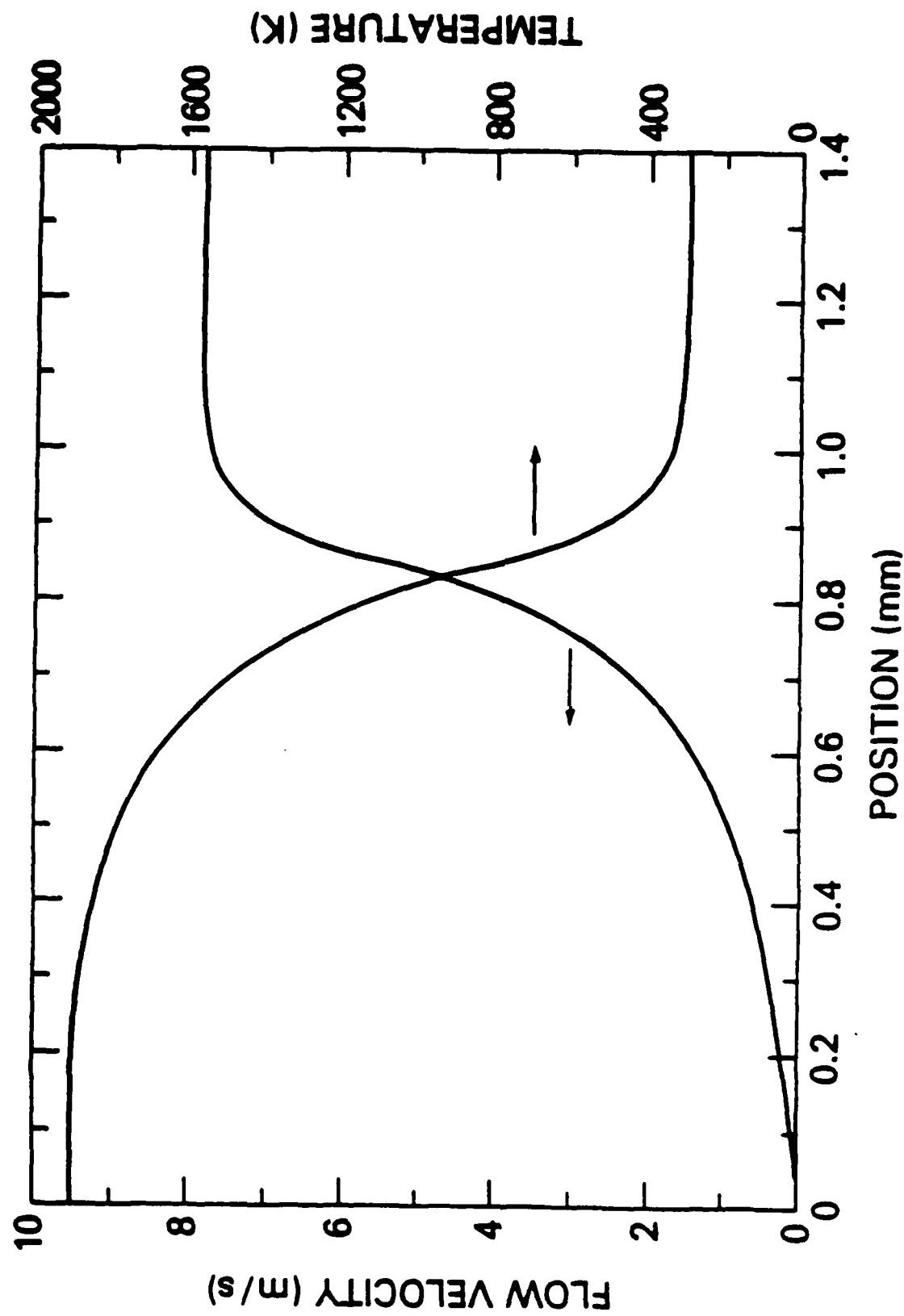


Fig. 1 — Velocity and temperature profiles in a planar flame propagating in an $\text{H}_2\text{:N}_2/2:14$ mixture.

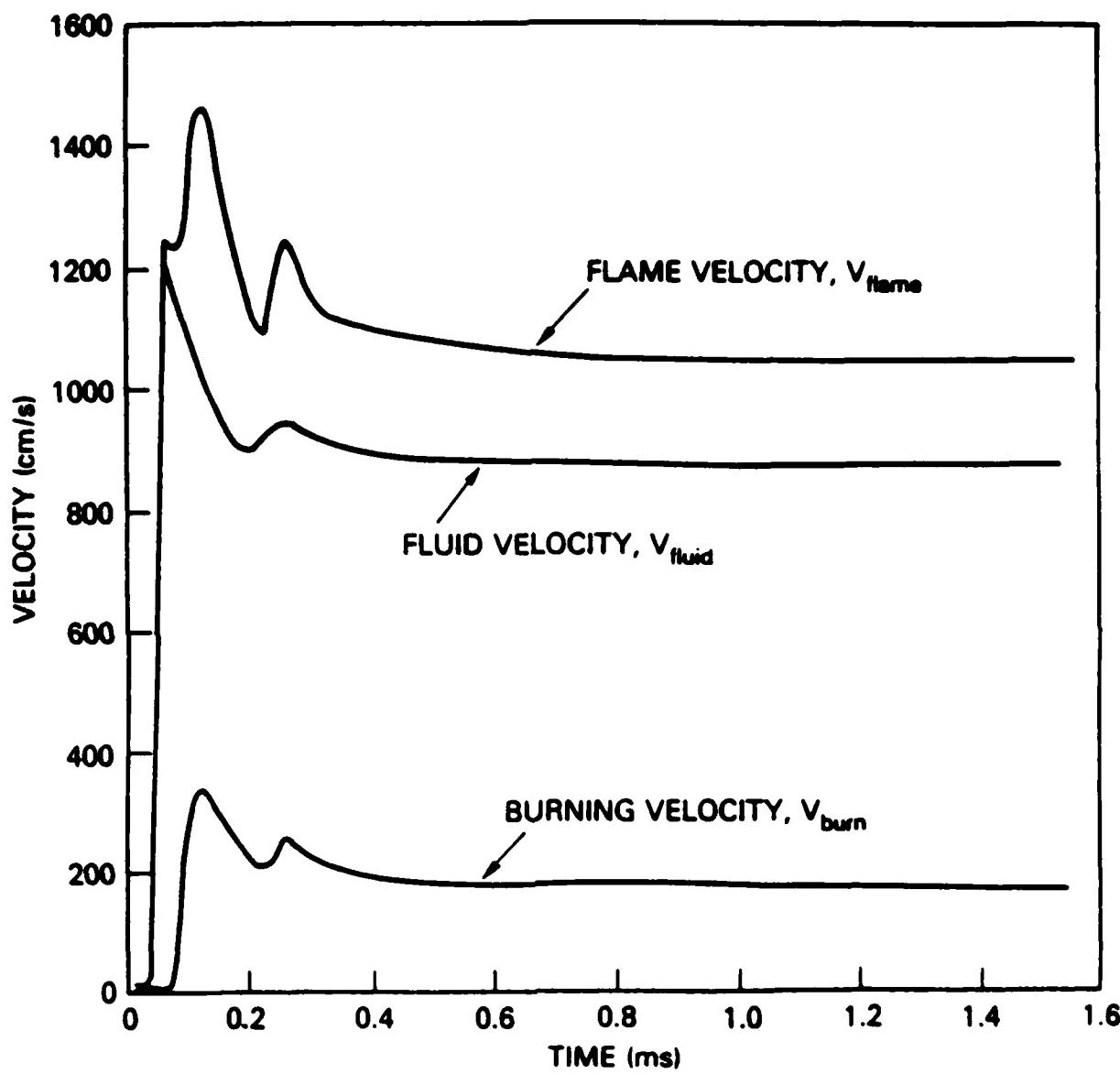


Fig. 2 — Calculated fluid, flame, and burning velocities as a function of time
for the mixture $\text{H}_2:\text{O}_2:\text{N}_2/2:1:3.76$ at 298 K and 1 atm.

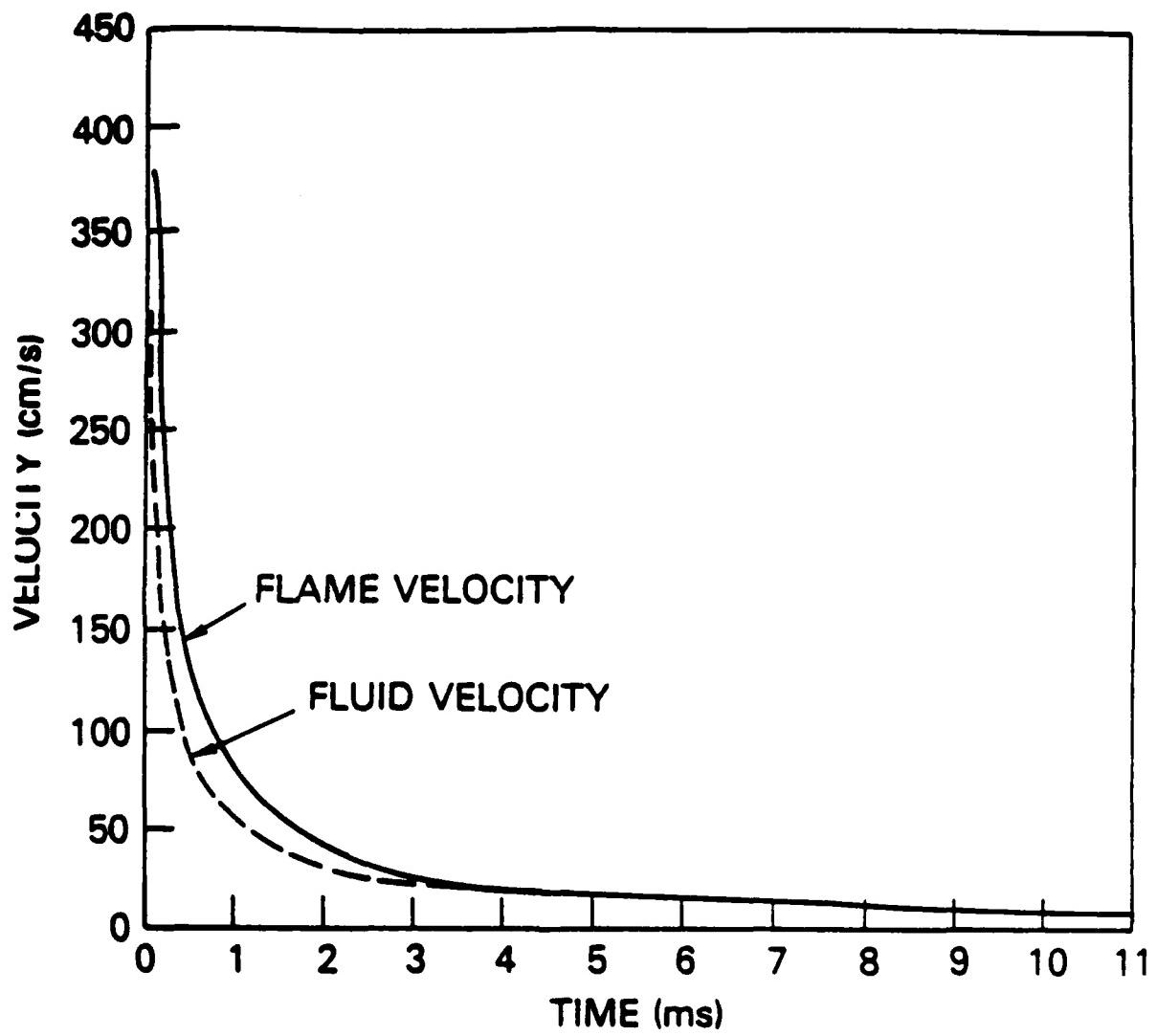


Fig. 3 — Calculated flame and fluid velocities as a function of time
for the $\text{H}_2:\text{O}_2:\text{N}_2/0.65:1:3.76$ mixture.

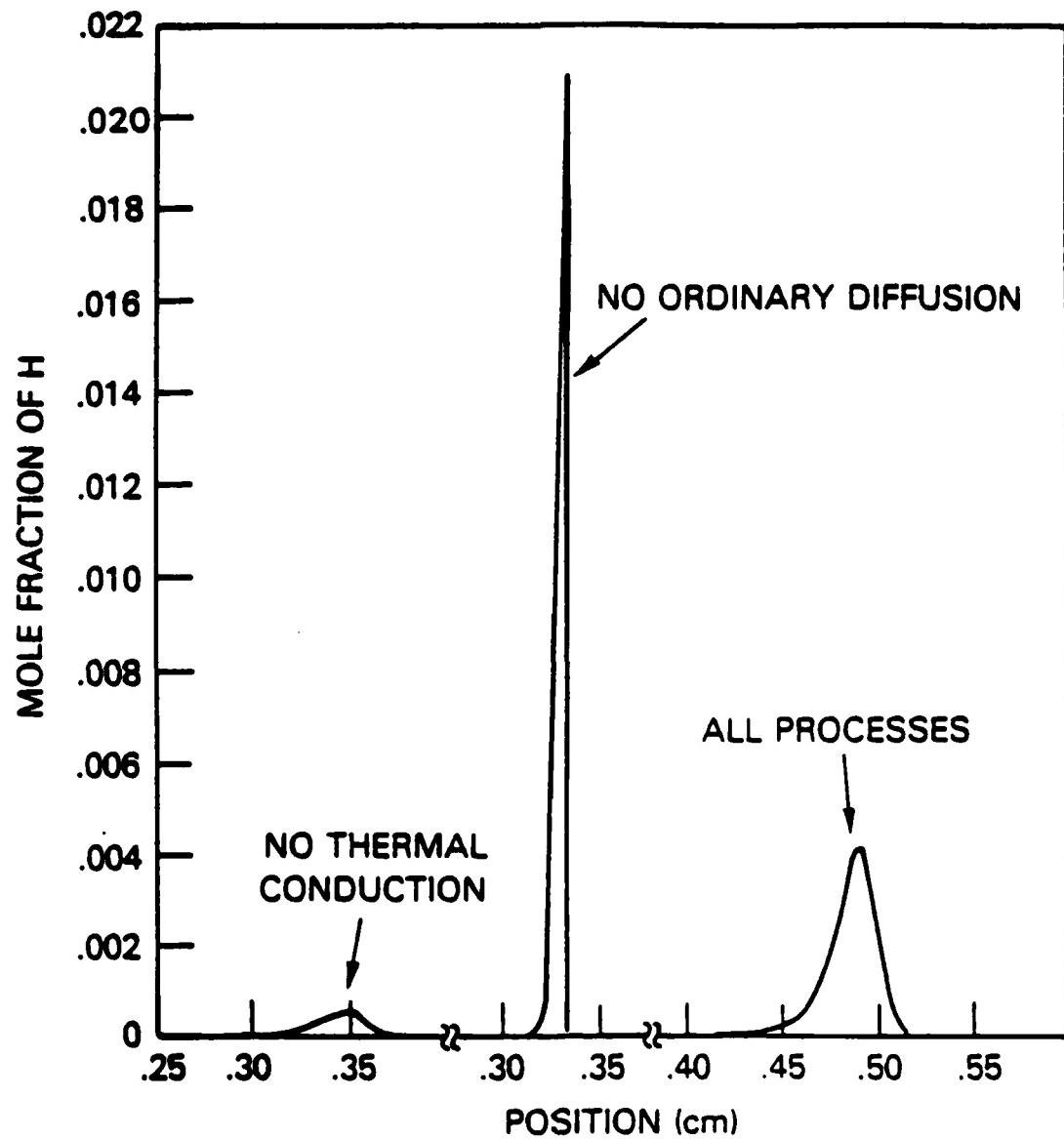


Fig. 4 — Hydrogen radical number densities at typical times in the calculation for the lean $H_2:O_2:N_2/0.65:1:3.76$ mixture.

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